

What Is Claimed Is:

1. Spherical silica-titania-based fine particles surface-treated with silane,  
 wherein a titanium atom content is within a range from 0.001 to 5% by weight, frictional  
 5 electrification with iron powder is within a range from -100 to -300  $\mu\text{C/g}$ , bulk density is  
 within a range from 0.2 to 0.4 g/ml, and particle diameter is within a range from 0.01 to 5  
 $\mu\text{m}$ .

2. A production process for the spherical silica-titania-based fine particles  
 10 surface-treated with silane according to claim 1, comprising the steps of  
 (A) producing hydrophobic spherical silica-titania fine particles by introducing  
 $\text{R}^5\text{SiO}_{3/2}$  units [wherein,  $\text{R}^5$  represents a substituted or unsubstituted monovalent  
 hydrocarbon group of 1 to 20 carbon atoms] onto a surface of hydrophilic spherical  
 silica-titania fine particles comprising  $\text{SiO}_2$  units and  $\text{TiO}_2$  units, and  
 15 (B) introducing  $\text{R}^7_3\text{SiO}_{1/2}$  units [wherein, said  $\text{R}^7$  groups are either identical or  
 different, and each represent a substituted or unsubstituted monovalent hydrocarbon  
 group of 1 to 6 carbon atoms] onto a surface of said hydrophobic spherical silica-titania  
 fine particles.

20 3. The production process according to claim 2, wherein said hydrophilic  
 spherical silica-titania fine particles used in said step (A) are produced by a process that  
 comprises a step for hydrolyzing and condensing a mixture of:

a tetrafunctional silane compound represented by a general formula (1):

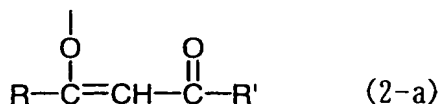


25 [wherein, said  $\text{R}^1$  groups are either identical or different, and each represent a  
 monovalent hydrocarbon group of 1 to 6 carbon atoms], or a partial  
 hydrolysis-condensation product thereof, or a mixture of the two; and

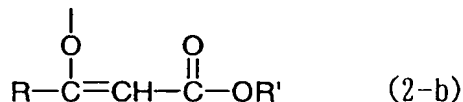
a tetrafunctional titanium compound represented by a general formula (2):



30 [wherein, said  $\text{R}^2$  groups are either identical or different, and each represent a  
 monovalent hydrocarbon group of 1 to 20 carbon atoms,  $\text{R}^3$  is either an enol residue of a  
 $\beta$ -diketone represented by a general formula (2-a):



(wherein, R and R' are either identical or different, and each represent a monovalent hydrocarbon group of 1 to 6 carbon atoms), or an enol residue of a  $\beta$ -ketoester represented by a general formula (2-b):



(wherein, R and R' are either identical or different, and are each as defined above), and p is an integer of 0 to 2] in a mixed liquid of water and a hydrophilic organic solvent in the presence of a basic material, thereby generating said hydrophilic spherical silica-titania fine particles.

4. The production process according to claim 3, wherein said tetrafunctional silane compound represented by the general formula (1) is selected from the group consisting of tetramethoxysilane, tetraethoxysilane, tetraisopropoxysilane, tetrabutoxysilane, and tetraphenoxysilane.

5. The production process according to claim 3, wherein said tetrafunctional silane compound represented by the general formula (2) is selected from the group consisting of titanium tetraisopropoxide, titanium tetrabutoxide, titanium tetrakis(2-ethylhexyloxide), titanium tetranonyloxide, titanium tetrastearoyloxide, titanium tetraisopropenoxide, titanium diisopropoxide bis(2,4-pentanedionate), titanium dibutoxide bis(2,4-pentanedionate), titanium diisopropoxide bis(2,2,6,6-tetramethyl-3,5-heptanedionate), and titanium diisopropoxide bis(ethylacetoacetate).

6. The production process according to claim 3, wherein said hydrophilic organic solvent is an alcohol solvent represented by a general formula (3):



[wherein, R<sup>4</sup> is a monovalent hydrocarbon group of 1 to 6 carbon atoms].

7. The production process according to any one of claims 3 to 6, wherein said basic material is ammonia.

5           8. The production process according to any one of claims 3 to 6, wherein the quantity of water used is within a range from 0.5 to 5 mols per 1 mol of alkoxy groups within the tetrafunctional silane compound of the general formula (1) or the partial hydrolysis-condensation product thereof and the tetrafunctional titanium compound of the general formula (2), and the ratio between the water and the hydrophilic organic  
10 solvent is a weight ratio within a range from 0.5 to 10.

          9. The production process according to any one of claims 3 to 6, wherein the quantity of the basic material is within a range from 0.01 to 5 mols per 1 mol of alkoxy groups within the silane compound of the general formula (1) or the partial  
15 hydrolysis-condensation product thereof and the compound of the general formula (2).

          10. The production process according to any one of claims 2 through 9, wherein in said step (A), a trifunctional silane compound represented by a general formula (4):



20 [wherein  $\text{R}^5$  is a substituted or unsubstituted monovalent hydrocarbon group of 1 to 20 carbon atoms, and said  $\text{R}^6$  groups are either identical or different, and each represent a monovalent hydrocarbon group of 1 to 6 carbon atoms], or a partial hydrolysis-condensation product thereof, or a mixture of the two is added to either an aqueous dispersion, or a mixed solvent dispersion of water and a hydrophilic organic  
25 solvent in the presence of said hydrophilic spherical silica-titania fine particles, thereby treating a surface of said hydrophilic spherical silica-titania fine particles and generating hydrophobic spherical silica-titania fine particles.

          11. The production process according to claim 10, wherein the trifunctional  
30 silane compound represented by the general formula (4) is selected from the group consisting of methyltrimethoxysilane, methyltriethoxysilane, ethyltrimethoxysilane, ethyltriethoxysilane, n-propyltrimethoxysilane, n-propyltriethoxysilane, isopropyltrimethoxysilane, isopropyltriethoxysilane, butyltrimethoxysilane,

butyltriethoxysilane, hexyltrimethoxysilane, trifluoropropyltrimethoxysilane and heptadecafluorodecyltrimethoxysilane, and a partial hydrolysis-condensation product of these compounds.

5           12. The production process according to claim 10, wherein the quantity of the trifunctional silane compound represented by the general formula (4) is within a range from 0.001 to 1 mol per 1 mol of combined SiO<sub>2</sub> units and TiO<sub>2</sub> units within the hydrophilic spherical silica-titania fine particles

10           13. The production process according to claim 10, wherein in said step (B), a dispersion medium of said aqueous dispersion comprising hydrophobic spherical silica-titania fine particles is replaced with a ketone-based solvent, thereby generating a ketone-based solvent dispersion comprising said hydrophobic spherical silica-titania fine particles, and either a silazane compound represented by a general formula (5):



[wherein, said R<sup>7</sup> groups are either identical or different, and each represent a substituted or unsubstituted monovalent hydrocarbon group of 1 to 6 carbon atoms], a monofunctional silane compound represented by a general formula (6):



20 [wherein, said R<sup>7</sup> groups are either identical or different and are as defined above, and X represents either an OH group or a hydrolysable group], or a mixture of the two is added to said ketone-based solvent dispersion comprising said hydrophobic spherical silica-titania fine particles, thereby triorganosilylating residual reactive groups at a surface of said hydrophobic spherical silica-titania fine particles.

25           14. The production process according to claim 13, wherein the quantity of the ketone-based solvent, reported as a weight ratio relative to the quantity of hydrophilic spherical silica-titania fine particles used, is within a range from 0.5 to 5.

30           15. The production process according to claim 13, wherein said ketone-based solvent is methyl isobutyl ketone.

16. The production process according to claim 13, wherein the silazane

compound represented by the general formula (5) is hexamethyldisilazane or hexaethyldisilazane.

17. The production process according to claim 13, wherein the monofunctional silane compound represented by the general formula (6) is selected from the group consisting of trimethylsilanol, triethylsilanol, trimethylchlorosilane, triethylchlorosilane, trimethylmethoxysilane, trimethylethoxysilane, trimethylsilyldimethylamine, trimethylsilyldiethylamine, and trimethylacetoxysilane.

18. The production process according to claim 13, wherein the quantity of the silazane compound, the monofunctional silane compound, or a mixture of the two is within a range from 0.1 to 0.5 mols per 1 mol of combined  $\text{SiO}_2$  units and  $\text{TiO}_2$  units within the hydrophilic spherical silica-titania fine particles.

19. An external additive for an electrostatically charged image developing toner comprising the spherical silica-titania-based fine particles surface-treated with silane according to claim 1.

20. The external additive for an electrostatically charged image developing toner according to claim 19, wherein the quantity of the external additive added to the toner is within a range from 0.01 to 30 parts by weight per 100 parts by weight of the toner.